

Response Reviewer 1. os-2017-32 Lizotte et al.

1. The methodology used is correct and well described. As a general comment, the only weakness detected on this study is that not all pools of DMS(P) cycling were covered since no measurements of DMSO were performed (particulate and dissolved) which hampers a more extended discussion on the fate of metabolized DMS in seawater.

Answer ML. Indeed, DMSO measurements would have been a very appreciable addition to the paper. Unfortunately, they were not available, and we therefore cannot rule on the fate of certain pools.

2. It is really appreciated negative results of influence of light preincubations on DMSP dynamics. I think it is not stressed enough in the discussion of the paper. One thinks it is a pity than in such DMSP-active zone more specific experiments to test still open questions of the cycle, mainly related to the different physiological and ecological roles of DMSP in the upper ocean could have been tested (for instance, the relative role of non-DMSP-producers algae as sink of DMSP, algal DMS production, new in situ production of DMSP by heterotrophic bacteria, chemotaxis, etc). Rather than a weakness, I hope the paper will encourage the DMSP community to sample in the described area.

Answer ML. We agree with the reviewer. We furthered the discussion by adding some information in the methodology section (lines 334-348), please see point#11 of this review for the full description of the added information.

3. line 38: there is more than only 2 fates of consumed DMSP, excretion as an oxidized form but not incorporated into cell structure is missed.

Answer ML. To address this we changed the phrase: “This study focused on the two opposing fates of DMSP-S following its uptake by microbial organisms: either its conversion into DMS, or its assimilation into bacterial biomass.” For the following: “This study focused **on two opposing short-term** fates of DMSP-S following its uptake by microbial organisms: either its conversion into DMS, or its assimilation into bacterial biomass.”

Then we also added information about the third fate in the introduction section: **“Another potential fate for DMSP is its transformation into dissolved non-volatile degradation products (DNVS), including sulfate (SO_4^{2-}), however less is known of the molecular pathways involved in this process (Kiene et al. 2000; Reisch et al. 2011).”**

4. line 45. "measured in this study" can be deleted.

Answer ML. The words “...measured in this study...” have been deleted.

5. line 59: Since no aerosols were measured, I wouldn't mention it in the abstract of the paper

Answer ML. The following phrase: “The findings from this study provide crucial information on the distribution and cycling of DMS and DMSP in a critically under-sampled area of the global ocean, and they highlight the importance of oceanic fronts as

hotspots of the production of marine biogenic S compounds and as potential sources of aerosols particularly in regions of low anthropogenic perturbations such as the frontal waters of the Southern Hemisphere.”, was changed to: “The findings from this study provide crucial information on the distribution and cycling of DMS and DMSP in a critically under-sampled area of the global ocean, and they highlight the importance of oceanic fronts as hotspots of the production of marine biogenic S compounds.”

6. line 70: *Quinn and Bates 2011 should be also cited since evidence for climate regulation though DMS still needs to be proven.*

Answer ML. The following phrase: “DMS has gained notoriety over several decades of research on the grounds of its potential role linking ocean biology and the climate (Andreae et al., 1985; Charlson et al., 1987; Lovelock et al., 1972).” Was changed to: “DMS has gained notoriety over several decades of research on the grounds of its potential role linking ocean biology and the climate (Andreae et al., 1985; Charlson et al., 1987; Lovelock et al., 1972), **a role that is still under debate (Quinn and Bates 2011, Quinn et al. 2017).**”

7. line 92: *misplacement of the (*

Answer ML. The parenthesis in the following phrase conforms to the requirements: “These productive regions sometimes form unique biogeographic habitats of their own such as the Subtropical Convergence province proposed by Longhurst (2007).”

8. line 149: *the sentence should read "...the potential climatic relevant gas..."*

Answer ML. The following phrase: “Depending on bacterial requirements for either S or C and the relative contribution of DMSP to the overall oceanic S pool (Kiene et al. 2000; Levasseur et al 1996; Pinhassi et al. 2005), at least two very different and competing outcomes are involved from the bacterial catabolism of DMSP: one producing DMS, the climatic relevant gas, the other producing methanethiol (MeSH), an important microbial substrate (Kiene and Linn, 2000b).”, was changed to: “Depending on bacterial requirements for either S or C and the relative contribution of DMSP to the overall oceanic S pool (Kiene et al. 2000; Levasseur et al 1996; Pinhassi et al. 2005), at least two very different and competing outcomes are involved from the bacterial catabolism of DMSP: one producing DMS, the **potential** climatic relevant gas, the other producing methanethiol (MeSH), an important microbial substrate (Kiene and Linn, 2000b).

9. line 218: *were the samples fixed with any fixative? P+G?*

Answer ML. No paraformaldehyde nor glutaraldehyde were used, rather the samples were snap-frozen in liquid N₂ and quickly analyzed after that. The following phrase: “Bacterial samples were frozen in liquid nitrogen (Lebaron et al., 1998) and thawed immediately before counting by flow cytometry following the methods described in Safi et al. (2007).” Was changed to: Bacterial samples were **snap-frozen** in liquid nitrogen (Lebaron et al., 1998) and thawed immediately before counting by flow cytometry shortly after the cruise following the methods described in Safi et al. (2007).”, in order to make it clearer.

10. line 221: *Dinoflagellate abundance was determined?*

Answer ML. Dinoflagellate abundance was determined in surface samples for all stations but not systematically for the “near surface” samples from which the incubation experiments were derived in this paper. It is thus possible to provide some information about the overall “regional” conditions of phytoplankton dominance shown in Table 1 but not to discuss the specific near surface abundances. The phytoplankton speciation data will be discussed in a separate DMS/marine biogeochemistry paper. Nevertheless we modified the information by adding a phrase: “Coccolithophore abundance **in near surface waters** was determined using optical microscopy as described in Chang and Northcote (2016).” **Dinoflagellate abundance was determined for surface waters (not for near surface waters) and is not shown here.”**

11. lines 314-325: *Very interesting results that can be more discussed after Ruiz-Gonzalez et al. ISME Journal (2012) 6, 650–65, for instance.*

Answer ML. It is true that the absence of a significant difference between pre-incubation treatments is interesting in itself. We added some discussion on this, referring to Ruiz-González et al. (2012) and other publications (specifically related to the sulfur-relevant responses) but also more particularly to the review published by Ruiz-González et al in 2013 which clearly shows that the past 20 years of research on sunlight-bacteria interactions display a wide-range in responses (from negative to positive effect of natural sunlight on metabolic activity of heterotrophic bacteria) intimately linked with factors such as the phylogeny of bacterial groups under investigation, the light-history experienced by the natural populations, and many more. The added information is in bold in the following section:

“On the whole, the light conditions (dark and ambient) at which the cells were pre-acclimated for 6 h had no significant effect on the ^{35}S -DMSP_d metabolic rates measured. **This result contrasts with findings from earlier studies (such as Galí et al., 2011; Ruiz-González et al., 2012a; Slezak et al., 2001, 2007; Toole et al., 2006) and could be related to a number of variables such as the timing and depth of sampling, the type of bacterial assemblages present and their previous light-history, as well as the different temporal and spatial scales at which exposure to solar radiation varies (Ruiz-González et al., 2013). Because of these wide-ranging and intricate light-bacteria interactions, natural solar radiation is believed to play a significant, yet challenging to predict, role in modulating bacterial dynamics and biogeochemical functions (Ruiz-González et al., 2013). In the current study, the sulfur-related metabolic activities of the marine biota sourced in the morning (between ca. 7h00 and 9h00; Table 1) from the highly irradiated near surface waters may have persisted in the dark within the time period of experimental pre-exposure (6 h), however the lack of information on the phylogeny of bacterial groups present, for example, hampers a more detailed discussion.** We therefore present rate measurements made in dark-incubated samples that had been pre-exposed to ambient light conditions for 6 h.”

12. line 448: *"Microbial affinity for DMSP_d as indicated by" can be deleted*

Answer ML. Yes. The following phrase: “Microbial affinity for DMSP_d, as indicated by the ^{35}S -DMSP_d loss rate constant (k_{DMSP_d} ; Fig. 3a) varied between 0.4 and 3.4 d⁻¹, with the exception of a higher value of 19.9 d⁻¹ measured in the B2 cluster at station 5.” Was

changed to: “The ^{35}S -DMSP_d loss rate constant (k_{DMSPd} ; Fig. 3a) varied between 0.4 and 3.4 d⁻¹, with the exception of a higher value of 19.9 d⁻¹ measured in the B2 cluster at station 5.”

13. line 651: *I love Table 3*

14. line 665: *Could cyanobacteria be included? Were them measured by flow cytometry? It is a pity no taxonomical description of the communities could be performed.*

Answer ML. Yes it is indeed a good idea to mention cyanobacteria here as they have been shown (particularly *Synechococcus* and *Prochlorococcus*) to participate in DMSP assimilation. The following phrase: “It has been suggested that loss rate constants of DMSP_d, rather than being directly related to stocks of bacteria could be more related to bacterial community composition, and particularly the specific abundance of *Roseobacter*, a member of Alphaproteobacteria, and with Gammaproteobacteria (Royer et al., 2010), which are both significant contributors to DMSP metabolism (Malmstrom et al., 2004a, 2004b; Vila-Costa et al., 2007; Vila et al., 2004).”, was changed to: “It has been suggested that loss rate constants of DMSP_d, rather than being directly related to stocks of bacteria could be more related to bacterial community composition, and particularly certain members of Alphaproteobacteria, Gammaproteobacteria **as well as cyanobacteria**, that could all potentially represent significant contributors to DMSP metabolism (Malmstrom et al., 2004a, 2004b, 2005; Royer et al., 2010; Vila-Costa et al., 2007; Vila et al., 2004). The appropriate references were also added (Vila-Costa et al. 2006a as well as Malmstrom et al. 2005). In reference to the other questions: we agree, it is highly unfortunate that no taxonomical description is available for the heterotrophic bacteria and picoplankton communities. This also limits our comprehension of the response of the biotic community under the different pre-incubation light exposure scenarios.

15. line 748: *What about the role of algal oxidative stress? do you have any measurement indicating senescence of the bloom during the sampled period of time?*

Answer ML: Measurements of photosynthetic efficiency (F_v/F_m) would have indeed been appreciable here, but are unfortunately not available. However we modified the phrase to reflect this possibility. The following phrase: “Community DMS production may have included indirect processes such as zooplankton grazing, viral lysis, and senescence, as well as direct algal DMSP-lyase activity associated with the presence of certain species of dinoflagellates and coccolithophores (Niki et al., 2000; Wolfe and Steinke, 1996), ubiquitous in Subantarctic waters in early March.”, was changed to: “Community DMS production may have included indirect processes such as zooplankton grazing, viral lysis, and senescence, as well as direct algal DMSP-lyase activity associated with the presence of certain species of dinoflagellates and coccolithophores (Niki et al., 2000; Wolfe and Steinke, 1996), ubiquitous in Subantarctic waters in early March, **and potential algal oxidative stress associated to light or nutrient availability (Stefels et al., 2007; Sunda et al., 2002).**

16. line 789: *"much needed" can be deleted.*

Answer ML: The phrase “Our study provides much needed information on both concentrations and cycling of dimethylated sulfur compounds within waters of the New Zealand biogeochemical province (NEWZ) and more specifically in an oceanic frontal region.” Was changed to: “Our study provides information on both concentrations and cycling of dimethylated sulfur compounds within waters of the New Zealand biogeochemical province (NEWZ) and more specifically in an oceanic frontal region.”

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The manuscript reports on measurements of dimethyl sulfur compounds DMSC (DMS and DMSP) concentrations and their cycling rates on both sides of the Subtropical Front near New Zealand. The study is part of the SOAP experiment and intends to relate DMSPC dynamics to hydrographic and biological characteristics. To do so, measurements concentrate in three different areas that are investigated with a Lagrangian approach. The DMSP availability hypothesis is used as the major driver for the interpretation of most of the data, yet with uneven fit. The authors conclude that, as previously suggested, oceanic fronts generate hotspots for the production and emission of dimethyl C1 sulfur. Even though no great advances in knowledge are provided that can be of applicability to a broad range of regions of the global ocean, the study is timely and the data valuable. The manuscript is well written and properly contextualized and referenced. I do not have major concerns towards publication but provide here below some questions and suggestions that may help improve the robustness and argumentation.

1. Methods, equation 1 and L206-213, also L541-550: SRD is calculated from dailyaveraged irradiance. Is it taken for the 24 hours prior to sampling? Or is it the 24 hours of the sampling day? The rationale of the SRD concept related to DMS (as from Vallina & Simó 2007) relies on the previous 24 hours, which is the time over which photobiological and photochemical processes led to the observed DMS concentration.

Answer ML. The calculations are indeed based on the daily irradiance averaged over the 24 hours prior to sampling. We made this clearer in the methodology by modifying the following sentence: “Solar radiation dose (SRD in W m^{-2}) was calculated using Eq. (1) where I_0 represents the daily-averaged irradiance (in W m^{-2}) measured using an Eppley Precision Spectral Pyronometer (285-2800 nm), k (in m^{-1}) are estimates of vertical diffuse attenuation coefficients based on Photosynthetically Active Radiation (PAR) offset between two depths (2 m and 10 m), MLD is the mixed layer depth defined as the point at which a 0.2°C difference from the sea surface temperature occurred and was calculated according to Kara et al. (2000).”, and changing it to: “Solar radiation dose (SRD in W m^{-2}) was calculated using Eq. (1) where I_0 represents the daily-averaged irradiance **of the 24 hours prior to sampling** (in W m^{-2}) measured using an Eppley Precision Spectral Pyronometer (285-2800 nm), k (in m^{-1}) are estimates of vertical diffuse attenuation coefficients based on Photosynthetically Active Radiation (PAR) offset between two depths (2 m and 10 m), MLD is the mixed layer depth defined as the point at which a 0.2°C difference from the sea surface temperature occurred and was calculated according to Kara et al. (2000).

2. L241-258: Provide details of how ^{35}S -DMSPd loss was measured – I guess it was by removal of ^{35}S -DMS, transformation of all the remaining ^{35}S -DMSPd into ^{35}S -DMS, which is trapped onto H_2O_2 -soaked filter. Am I right?

Answer ML. The ^{35}S -DMSP_d loss rate is measured by the disappearance of dissolved ^{35}S -DMSP over time: the loss of ^{35}S -DMSP_d reflecting what is being consumed. To add clarity to this part of the paper we included more information by modifying the following sentences: “The bottles were then incubated for 3 h at *in situ* temperature during which time subsamples were taken after 0, 30, 60, and 180 min to measure the loss of ^{35}S -DMSP_d over time. The k_{DMSPd} was calculated as the slope of the natural log of the fraction of remaining ^{35}S -DMSP_d versus time.” to these ones: “The bottles were then incubated for 3 h at *in situ* temperature during which time **1 mL** subsamples were taken after 0, 30, 60, and 180 min **and transferred into 10-mL scintillation vials containing 5 mL Ecolume™ in order** to measure the loss of ^{35}S -DMSP_d over time **(the disappearance of ^{35}S -DMSP_d representing the consumption of this pool)**. The k_{DMSPd} was then calculated as the slope of the natural log of the fraction of remaining ^{35}S -DMSP_d versus time.”

The “transformation of all the remaining ^{35}S -DMSPd into ^{35}S -DMS, which is trapped onto H_2O_2 -soaked filter” mentioned by the reviewer is called the “unreacted or unconsumed dissolved ^{35}S -DMSP” which was measured at the end of the incubation period. We discuss this around lines 279-283: “After the volatiles were trapped, a new stopper with H_2O_2 -soaked filter was placed in the vial. Each vial was then injected with 0.2 mL NaOH (5N) through the stopper using a BD precision guide needle to quantitatively cleave remaining ^{35}S -DMSP_d into ^{35}S -DMS. The ^{35}S -DMS was trapped as described above.” To make it clearer what this pool represents we modified the phrase which now reads as follows: “After the volatiles were trapped, a new stopper with H_2O_2 -soaked filter was placed in the vial. Each vial was then injected with 0.2 mL NaOH (5N) through the stopper using a BD precision guide needle to quantitatively cleave the remaining ^{35}S -DMSP_d into ^{35}S -DMS **(a pool known as the unconsumed ^{35}S -DMSP_d)**. The ^{35}S -DMS was trapped as described above.”

3. L341-342: How was the cryogenic trap cooled to -20°C ?

Answer ML. The trap was encased in a metal block that also contained a cold finger connected to an external cryo-cooling unit monitored and controlled automatically. The following phrase: “Briefly, calibrated volumes (5 mL) of seawater samples were purged with zero-grade nitrogen (99.9 % pure) and gas-phase DMS was cryogenically concentrated on 60/80 Tenax TA in a stainless steel trap at -20°C , then thermally desorbed at 100°C for analysis by GC coupled with sulfur chemiluminescent detection.”, was changed to: “Briefly, calibrated volumes (5 mL) of seawater samples were purged with zero-grade nitrogen (99.9 % pure) and gas-phase DMS was cryogenically concentrated on 60/80 Tenax TA in a stainless steel trap **maintained at -20°C via a cold finger connected to a cryo-cooling unit**, then thermally desorbed at 100°C for analysis by GC coupled with sulfur chemiluminescent detection.”

4. Results, L464-466: *A bacterial DMS production rate (from DMSP_d only) of 27 nmol/Ld is astonishingly high, more so when DMS concentration is 3 nmol/L and DMSP_d is 10 μm, namely dinoflagellates.*

Answer ML. We agree with the reviewer, this rate is quite high. Such high rates are rare but have been published before (Royer et al. 2010). We are confident however that this is not a problem with a specific incubation (or bottle effect) since all the incubations displayed the same results (even the duplicate dark-acclimated samples that we do not present in the paper, as mentioned in the methodology section, were extremely high and showed no significant differences with the light-acclimated samples that we discuss in the paper). We added the following phrases to this section in order to reflect potential reasons for this response: **“This high rate reflects the very high DMSP_d scavenging by the bacteria measured on this particular day. The fact that concentrations of DMS remained low (ca. 3 nmol L⁻¹) suggests that potential sinks, particularly bacterial DMS consumption, but not excluding DMS photo-oxidation and ventilation (Table 1) may have kept this pool in check.”**

5. Figure 5: *Correlation between DMSP_t and chlorophyll *a* is quite strong indeed. One would expect it even stronger with DMSP_p, since it is better associated with algal cells. Perhaps it does not deserve another graph but some mention to the regression facts.*

Answer ML. The strength of the regression between DMSP_p and Chl *a* ($r^2 = 0.57$) is very similar to the one between DMSP_t and Chl *a* ($r^2 = 0.59$). We added this information in the discussion section (starting at line 541): **“A type II linear regression model suggests that 59% of the variance in pools of DMSP_t can be explained by the variability in stocks of chl *a* (Fig. 5a), while the strength of the relationship between DMSP_p and chl *a* is also strong ($r^2 = 0.57$, data not shown).”**

6. C3 Table 1: *All variables are reported “in blooms” and in the vicinity (N or S of). But chlorophyll concentrations are not any lower in the vicinities. So, what is the definition of bloom?*

Answer ML. This is discussed in detail by the SOAP overview paper (Law et al. also currently under review). However to add some precision to this aspect we added the following phrase to the methodology section (lines 204-209). **“The SOAP blooms were coherent discrete areas of elevated ocean colour identified in satellite images characterised by a maximum of 1 mg/m³ chl *a* or higher. Sampling took place near the center of these blooms but also at stations on the periphery and outside the blooms (Table 1), as defined by the distance from the bloom centre and clear demarcation in surface biogeochemical variables (see Law et al., this issue).”**

In this paper, we separate the stations in “clusters” (see all figures and discussions ensuing), to account for the fact that stations are either directly “in” or “in the vicinity of” the blooms.

7. *Same for nutrients and DMSP:Chla.*

Answer ML. We are not certain what the reviewer is asking here. If possible, added information would help us address any concerns regarding this part of the paper. Thank you.

8. I like the data compilation in Table 3.

Answer ML. We thank both reviewers for acknowledging this positive aspect of the paper.